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was conducted

Rice University has conducted an investigation of the fundamental processes affecting the operation, performance and application of electrically excited lasers having potential utility in a variety of areas of importance to the Navy. Particular attention was focused on the broadband XeF(C→A) laser, which has the potential for efficient, tunable operation throughout the blue-green spectral region. The research at Rice University has been coordinated with a complementary ONR-supported experimental program conducted at United Technologies Research Center. This ~~Final~~ Report summarizes the results of this investigation, and lists published papers and conference reports in which specific results and conclusions of the research are described in detail. Keywords.

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STUDIES OF TUNABLE EXCIMER LASER SOURCES

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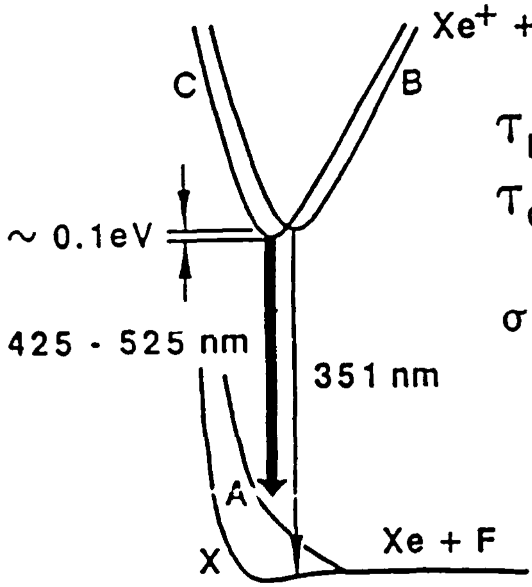
The principal area of research under this grant has been the development of tunable broadband excimer laser sources in the visible and UV spectrum, with emphasis on fundamental laser physics issues of the $XeF(C \rightarrow A)$ *excimer laser.*

The high efficiency, scalability to high powers and reliability of excimer lasers make them useful devices in an increasing number of scientific, industrial, and medical applications such as various types of spectroscopy, remote sensing, materials processing, and optical communications. Most excimer lasers in existence today involve a diatomic rare gas halide molecule that operates on relatively narrow band transitions in the UV region of the spectrum. The most important of these are the ArF laser at 193 nm, KrF at 248 nm, $XeCl$ at 308 nm, and XeF at 351 nm. These excimer lasers generally employ an electric discharge or electron beam to deposit energy into rare gas mixtures with a halogen containing fuel. The usefulness of these lasers could be greatly enhanced if excimer lasers could be tuned over broad wavelength intervals similar to dye or tunable solid state lasers.

Since 1979 there has been considerable progress in achieving this goal as a result of extensive studies of electron beam excited broadband diatomic and triatomic excimers at Rice University, and important early contributions by several other groups [1-11]. The Rice University group reported the first observation of three new electron beam pumped rare gas halide excimer lasers with wavelength tunability over the range from 415 to 540 nm. These are the $XeF(C \rightarrow A)$ laser centered at 485 nm [12], as well as the Xe_2Cl laser at 520 nm [13], and the Kr_2F laser at 435 nm [14]. Because they operate in the blue-green region of the spectrum, they have important potential applications. The most promising of this class of lasers is the $XeF(C \rightarrow A)$ laser with continuous tunability and narrowband laser output in the blue-green spectral range from 435 to 535 nm.

The basic physical properties of the $XeF(C \rightarrow A)$ system are outlined in Fig. 1. The $C \rightarrow A$ transition is a typical excimer bound-free transition with the lower potential curve being steeply repulsive. This causes a broad band emission ranging from approximately 420 nm to 540 nm, i.e., from the deep violet into the green. The radiative lifetime of the C states is 100 ns, which is almost an order of magnitude larger than the B-state lifetime. The large bandwidth, together with the smaller radiative transition probability, causes the cross section for stimulated emission of the $C \rightarrow A$ laser ($1 \cdot 10^{-17}\,cm^2$) to be about a factor of 30 smaller than for the comparable $B \rightarrow X$ transition. The XeF system is unique, however, in that the C state lies about 0.1 eV below the B-state, which facilitates the build-up of large populations in the C state.

This laser originally suffered from low efficiency (<<1%) when electrically excited, due primarily to the occurrence of severe transient broadband and narrowband absorptions by the laser medium. However, progress by the Rice University group, in close cooperation with W.L.



$$\tau_{B-X} \sim 15\ \text{ns}$$
$$\tau_{C-A} \sim 100\ \text{ns}$$
$$\sigma_s \sim \frac{\lambda^4}{\delta\lambda\tau} \begin{cases} B - X \sim 3.5 \times 10^{-16}\,cm^2 \\ C - A \sim 1.0 \times 10^{-17}\,cm^2 \end{cases}$$

Fig. 1: Potential diagram of the *XeF* system.

Nighan at United Technologies Research Center, has resulted in the identification of those species responsible for the transient induced absorptions [15]. Controlling these absorbers has led to significantly improved $XeF(C \rightarrow A)$ laser performance using electron-beam excitation of complex multi-component gas mixtures specifically tailored so as to reduce medium transient absorption in the blue/green region [16]. The use of Ar and Kr together as the effective rare gas buffer/energy transfer species, along with a combination of NF_3 and F_2 to produce the desired F-donor molecule characteristics, has permitted synthesis of near optimum medium properties for which $XeF(C)$ is produced efficiently while transient absorptions are minimized [17]. The use of such mixtures and injection control by means of an external dye laser has resulted in a laser output pulse energy density and intrinsic efficiency of ~2 J/liter and ≤2%, respectively. These values are comparable to those known for the rare gas halide $(B \rightarrow X)$ systems.

2. Efficient Wavelength Tuning of the XeF(C→A) Excimer Laser

The most remarkable property of the $XeF(C \rightarrow A)$ laser is its large wavelength tuning range of ~100 nm from 435 to 535 nm. Continuous tuning of the e-beam pumped laser, with a decrease in energy by less than 50%, has been demonstrated over a range of ~50 nm. It should be noted that the usable bandwidth from previous experiments was largely limited by the experimental apparatus (optics, etc.), and thus a larger portion of the fluorescence bandwidth may be accessible for amplification. The small cross section for the stimulated emission of the $(C \rightarrow A)$ transition enables most of the internal absorption to be bleached.

In order to obtain good beam quality, convenient tuning, and narrow linewidth on the $XeF(C \rightarrow A)$ laser transition, the electron beam pumped $XeF(C \rightarrow A)$ system was used as a regenerative amplifier for an injected dye laser pulse [18]. A high pressure multi-component gas mixture comprised of 1 Torr of F_2, 8 Torr of NF_3, 10 Torr of Xe, 300 Torr of Kr, and 6 atm of Ar was pumped by an electron beam (1 MeV, 260 A cm^{-2}, 10 ns FWHM). The deposited energy was 110 J/l in a cylindrical gain volume of 10 cm length and 2 cm diameter. For these conditions, the $XeF(C \rightarrow A)$ laser gain has a maximum value of ~3% cm^{-1} and a duration of ~35 ns (FWHM) [16]. A peak gain coefficient in excess of 2% cm^{-1} was found between 455 nm and 530 nm. A confocal unstable resonator of magnification 1.2, with a mirror distance of 12.5 cm was used to permit multiple-pass amplification of the injected pulse. A 250 ns long pulse from a coaxial flashlamp pumped dye laser was injected into the resonator through a central coupling hole of 1.5 mm diameter. The injected energy could be varied up to ~1 mJ in a 20 ns time window given by the temporal gain length of the $XeF(C \rightarrow A)$ amplifier. The long dye laser pulse provides a quasi - CW seed signal for the amplifier.

Presented in Fig. 2 are the measured energy extraction characteristics as a function of wavelength corresponding to an injection power density value of 1.1 MW/cm^2. This figure depicts the superposition of the spectra of twenty-three separate injection-controlled laser shots in the blue-green spectrum, and shows that with a relatively high injection power the valleys become very much less pronounced (red), and the wavelength tuning curve becomes relatively smooth. In fact, the output energy varied only by a factor of two over a 30 nm region (blue-red). This decrease in the induced transient absorption appears to result from bleaching by the intense injection photon flux. This is also confirmed by gain measurement as a function of probe laser power density. For the conditions depicted in Fig. 2, output energy density values between 0.7 and 1.3 J/l are obtained over a 50 nm wavelength range corresponding to an intrinsic efficiency of $\geq 1\%$. This figure also shows the spectrum of the free running laser, where the absorbers have not been saturated.

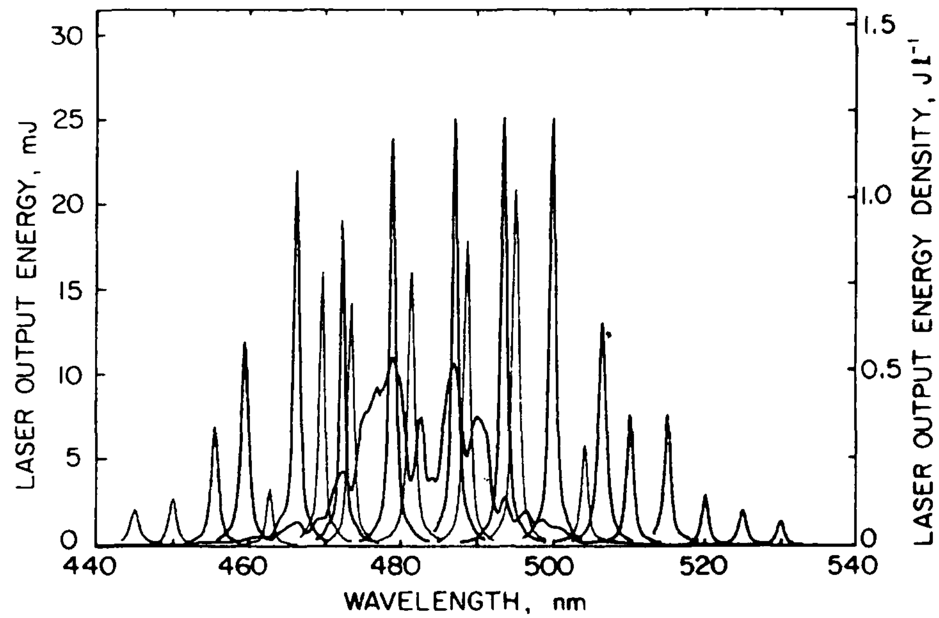


Fig. 2: Tuning spectrum for the *XeF(C→A)* laser. Amplified *XeF(C→A)* laser output for several separate injection-controlled shots is depicted. The narrowband absorptions can be bleached by the high photon flux leading to *continuous efficient* tuning over a large bandwidth. The amplified laser output for resonator optics centered at 480 nm is shown in blue and red, where the latter correspond to injection wavelengths set respectively at the peak and valley positions of the free running spectrum (black) at an injection power density of 1.1 MW/cm^2 (200 μJ in 10 ns, 1.5 mm beam diameter). The output shown in green corresponds to two further sets of cavity optics centered at 460 and 510 nm respectively.

3. Analytical Modeling of Injection Controlled Excimer Laser Amplifiers

A semi-empirical model of a pulsed, injection-controlled laser was developed and applied to the experimental results that have been obtained for the electron-beam pumped *XeF(C→A)* excimer laser. The gain medium inside an unstable cavity is represented by a folded pulsed amplifier which is seeded by a narrowband input signal [19]. A set of coupled rate equations for the population densities of the upper laser states, the wide-band absorbers, and the photon flux was numerically integrated. Measured gain and absorption of the amplifier were used as input data to evaluate the model. The model is used to predict the performance of the *XeF(C→A)* excimer laser over a wide range of experimental configurations using both internal and external optical resonators. Excellent agreement is achieved between mode predictions and experimental measurements of laser output energy and temporal laser profiles. Predicted and observed laser amplifier characteristics for different unstable resonator geometries, window losses, and cavity lengths could also be compared. This model also predicts the experimentally determined injection-control threshold for *KrF* excimer lasers successfully, and should be applicable to numerous other injection seeded lasers.

4. Simultaneous Multiwavelength Operation of Excimer Lasers

For applications in areas such as spectroscopy, optical diagnostics, and materials processing, it may be useful to have the capability of operating a single rare gas halide excimer laser system at two or more wavelengths. We demonstrated efficient simultaneous multiwavelength UV/visible operation of excimer lasers in both electron beam and discharge pumped laser systems.

It was found that an electron beam excited medium, which had been optimized for efficient blue/green $XeF(C \rightarrow A)$ laser oscillations, also exhibited strong net gain on the UV $B \rightarrow X$ transition, and that simultaneous laser oscillation on both transitions was possible. Subsequently, relatively efficient (0.25%) simultaneous UV/visible *XeF* laser oscillation was achieved through use of an optimized dual wavelength resonator along with the addition of *Kr* to the gas mixture. In this case the intensity of the 248 nm $KrF(B \rightarrow X)$ fluorescence was observed to be comparable to that of the 351 nm $XeF(B \rightarrow X)$ fluorescence, under conditions for which the dual $XeF(B \rightarrow X)$ and $(C \rightarrow A)$ laser output energies were equal. Since the kinetics of the $B \rightarrow X$ transitions of *KrF* and *XeF* are actually less competitive than those of the $XeF(B \rightarrow X)$ and $(C \rightarrow A$ transitions, this observation suggested that simultaneous multiple wavelength oscillation on $B \rightarrow X$ rare gas-halide transitions should be possible using appropriate gas mixtures in a commercial discharge excited excimer laser. Fig. 3 shows the time integrated UV/visible laser pulse energy density as a function of Kr pressure for representative conditions. As the Kr pressure is increased, the $XeF(C \rightarrow A)$ output decreased gradually in response to the increasing importance of competitive KrF and Kr_2F reactions, the later species is a strong absorber at 351 nm. Hence, the resultant increase in $XeF(C \rightarrow A)$ gain with increasing Kr pressure, combined with the decreasing competitive $B \rightarrow X$ oscillation, leads to a gradual increase in broadband $C \rightarrow A$ output centered at ~480 nm. The combined UV/visible output exceeded 0.5 J/l

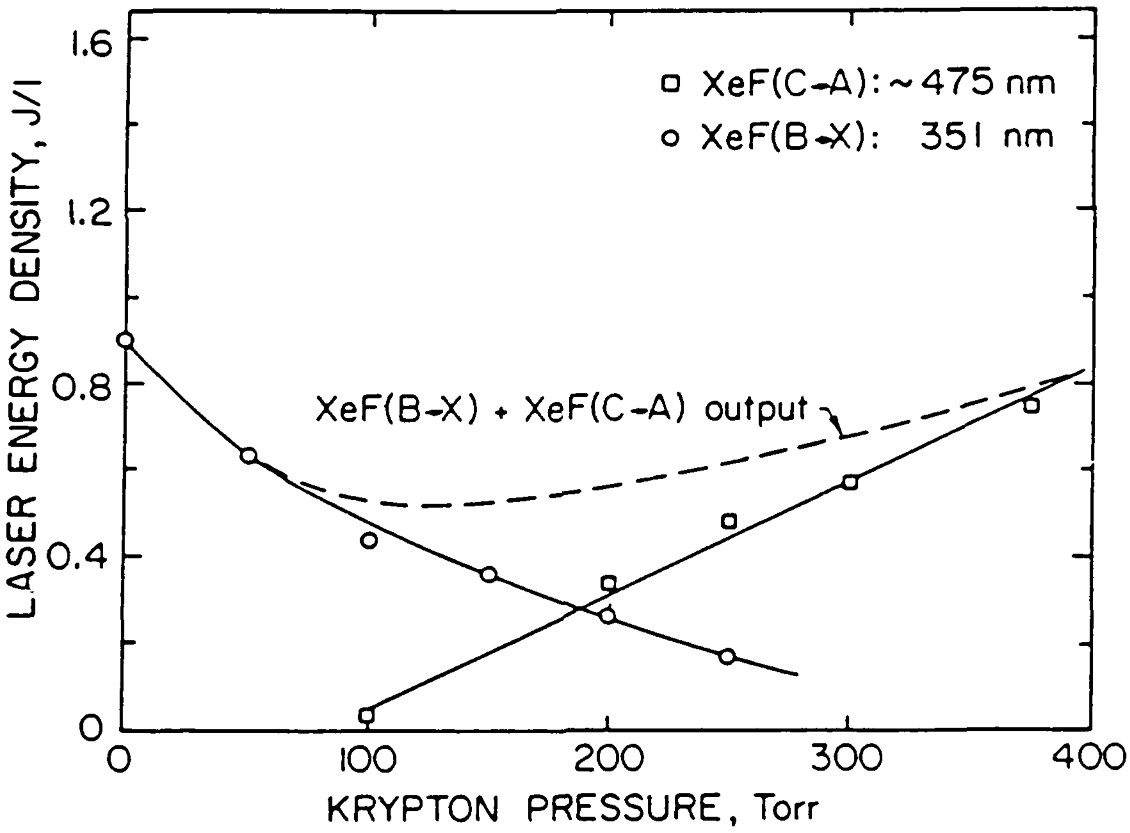


Fig. 3: Measured laser pulse energy density for the simultaneously occurring $B \rightarrow X$ and $C \rightarrow A$ XeF excimer transitions in an e-beam excited mixture comprised of 6.5 atm Ar, 8 Torr Xe, 8 Torr NF_3, 1 Torr F_2, and variable Kr pressure. The e-beam energy deposition was approximately 135 J/liter.

throughout the entire range of Kr pressures [20].

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Personnel Associated with Contract

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N. Hamada	Graduate Student
J. Hooten	Technical Staff
G. Marowsky	Consultant
W.L. Nighan	Consultant
N. Nishida	Graduate Student
M.C. Smayling	Graduate Student
F. Steigerwald	Research Assistant
H. Stiegler	Graduate Student
W. Walter	Research Assistant
Y. Zhu	Graduate Student

Honors / Awards / Prizes

IEEE Fellowship Award to F.K. Tittel

Optical Society of America Fellowship Award to F.K. Tittel

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